AFRL-RX-WP-JA-2016-0323

OPTICAL PROPERTIES OF LITHIUM TERBIUM FLUORIDE AND IMPLICATIONS FOR PERFORMANCE IN HIGH POWER LASERS (POSTPRINT)

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Northrop Grumman

16 October 2015
Interim Report

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MATERIALS AND MANUFACTURING DIRECTORATE
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AIR FORCE MATERIEL COMMAND
UNITED STATES AIR FORCE

REPORT DOCUMENTATION PAGE

Form Approved OMB No. 0704-0188

1. REPORT DATE *(DD-MM-YY)* 16 October 2015

2. REPORT TYPE Interim

3. DATES COVERED *(From - To)* 6 May 2010 – 16 September 2015

4. TITLE AND SUBTITLE
OPTICAL PROPERTIES OF LITHIUM TERBIUM FLUORIDE AND IMPLICATIONS FOR PERFORMANCE IN HIGH POWER LASERS (POSTPRINT)

5a. CONTRACT NUMBER IN-HOUSE

5b. GRANT NUMBER

5c. PROGRAM ELEMENT NUMBER

6. AUTHOR(S)
1) David E. Zelmon and Emily C. Erdman - AFRL/RX
2) Kevin T. Stevens, Greg Foundos, Joo Ro Kim, and Allen Brady – Northrop Grumman

5d. PROJECT NUMBER

5e. TASK NUMBER

5f. WORK UNIT NUMBER X09X

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
1) AFRL/RX
Wright-Patterson AFB, OH 45433
2) Northrop Grumman Synoptics,
1201 Continental Blvd, Charlotte, NC 28273

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
Air Force Research Laboratory
Materials and Manufacturing Directorate
Wright-Patterson Air Force Base, OH 45433-7750
Air Force Materiel Command
United States Air Force

10. SPONSORING/MONITORING AGENCY ACRONYM(S) AFRL/RXAP

11. SPONSORING/MONITORING AGENCY REPORT NUMBER(S) AFRL-RX-WP-JA-2016-0323

12. DISTRIBUTION/AVAILABILITY STATEMENT
Distribution Statement A. Approved for public release: distribution unlimited.

13. SUPPLEMENTARY NOTES
PA Case Number: 88ABW-2015-4958; Clearance Date: 16 Oct 2015. This document contains color. Journal article published in Applied Optics, Vol. 55, No. 4, 1 Feb 2016.
The final publication is available at http://dx.doi.org/10.1364/AO.55.000834

14. ABSTRACT *(Maximum 200 words)*
LiTbF4 has the potential to replace traditional magneto-optic (MO) garnet materials as a Faraday rotator in high power laser systems due to its high Verdet constant. New measurements are reported of the ordinary and extraordinary refractive indices of LiTbF4 as functions of wavelength and temperature respectively, as well as their corresponding Sellmeier expressions. Consequently, the Verdet coefficient was calculated and plotted as a function of wavelength and temperature. These measurements will aid in further development of LiTbF4 as an optical isolator.

15. SUBJECT TERMS
LiTbF4; magneto-optic (MO) garnet materials; Faraday rotator; high power laser; Verdet constant; Sellmeier; optical isolator

16. SECURITY CLASSIFICATION OF: a. REPORT	b. ABSTRACT	c. THIS PAGE	17. LIMITATION OF ABSTRACT:	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON (Monitor)
Unclassified	Unclassified	Unclassified	SAR	6	Steven Fairchild
					19b. TELEPHONE NUMBER *(Include Area Code)* (937) 904-4328

applied optics

Optical properties of lithium terbium fluoride and implications for performance in high power lasers

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Received 22 October 2015; revised 15 December 2015; accepted 21 December 2015; posted 22 December 2015 (Doc. ID 252482); published 1 February 2016

$LiTbF_4$ has the potential to replace traditional magneto-optic garnet materials as a Faraday rotator in high power laser systems due to its high Verdet constant. New measurements are reported of the ordinary and extraordinary refractive indices of $LiTbF_4$ as functions of wavelength and temperature, respectively, as well as their corresponding Sellmeier expressions. Consequently, the Verdet coefficient was calculated and plotted as a function of wavelength and temperature. These measurements will aid in further development of $LiTbF_4$ as an optical isolator.

OCIS codes: (160.3820) Magneto-optical materials; (160.4670) Optical materials; (230.3810) Magneto-optic systems.

http://dx.doi.org/10.1364/AO.55.000834

1. INTRODUCTION

Magneto-optical materials have been the subject of intense study for several decades. The independence of the polarization rotation on propagation direction makes magneto-optic devices ideal for a variety of applications including switching, modulation, interferometry, imaging, biomolecular detection, and optical isolation [1–5]. Faraday rotators are the basis for optical isolation and light amplitude modulation. For example, the most popular and principal technique in high power laser systems is linear polarization rotation by Faraday elements (FEs) for laser output extraction of the system. FEs are also used as isolators in laser chains and birefringence compensation in a solid-state laser medium [6]. For FEs being used in high power laser systems, a large Verdet constant, small absorption, small scattering losses, and a small nonlinear refractive index are necessary. These properties are characteristic of low-dispersion fluoride hosts, such as alkali fluorides [7], making them excellent choices for FEs. However, as the power of laser systems grows larger, FEs are exposed to very high field densities which can alter their optical and physical properties due to nonuniform temperatures in the element. In order to compensate for these changes, the temperature dependence of basic optical and mechanical properties such as the refractive index and stress-optic coefficients must be known. In this paper, we describe the measurement of the refractive index and its dependence on temperature of $LiTbF_4$.

2. EXPERIMENTS

Single crystals of $LiTbF_4$ were grown at Northrop Grumman SYNOPTICS by the Czochralski technique, utilizing an inert gas resistance furnace. The system is incongruently melting, with several reported peritectic compositions. A melt containing 63 mol. % LiF, the peritectic composition reported by Weber [3], was chosen as the starting composition. The melt was prepared using 4-9's purity TbF_3 and LiF, obtained from suppliers previously qualified by SYNOPTICS. A nitrogen atmosphere was employed for both the melting and growth at temperatures less than 900°C.

Crystals grown from the 63 mol. % LiF melt were just under 4 cm in diameter × 7 cm in length. These crystals had inclusions, as well as scattering centers (precipitates), but did contain areas of high quality material. Starting melt compositions (percent molar compositions) were varied systematically to improve crystal quality. However, more work is required to better understand the melt composition needed for high optical quality crystals.

The method of minimum deviation was used in order to obtain the refractive indices of the $LiTbF_4$ crystal, using the Moller–Wedel divided circle spectrometer [8]. $LiTbF_4$ is a tetragonal crystal with a 4/m point group and is therefore uniaxial. Triangular prisms of $LiTbF_4$ were cut from the boule with the optic axis perpendicular to the triangular faces. In this way, the ordinary and extraordinary indices could be measured

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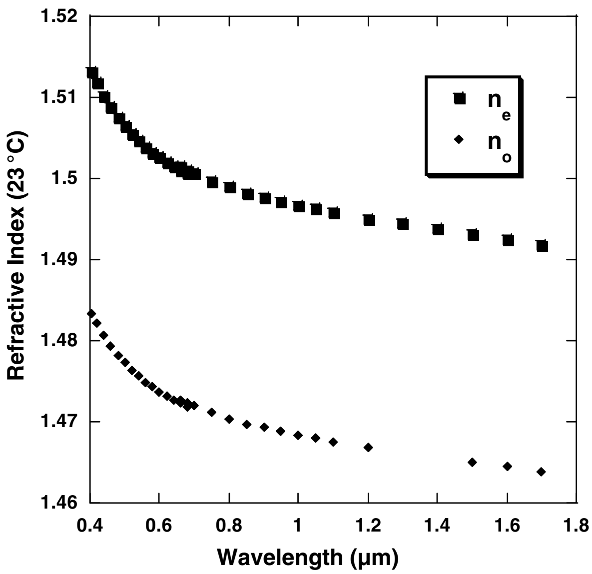

Fig. 1. Room temperature refractive indices of $LiTbF_4$.

independently by changing the polarization of the input light. To standardize the system, measurements of the refractive indices were made on a calcium fluoride prism from 0.4 to 5.0 μm. These measurements correlated with the published literature to within 0.0001 [9].

The apex angle was obtained by using an autocollimator attached to a Moller–Wedel divided circle spectrometer. The measured valued of the apex angle was 62.389° ± 0.002°. The tolerance on the measurements is the standard deviation calculated from 10 separate measurements of the apex angle.

Light produced by a mercury xenon source or an infrared source was coupled into a monochromator to provide discrete wavelengths ranging from 4 to 5.0 μm and then transmitted through a prism of $LiTbF_4$. Different detectors were used to detect the refracted light depending on the spectral range. This measurement was repeated five times and the average reported for each wavelength. The error estimated at any individual wavelength was less than 1.5×10^{-4}.

Measuring the change of refractive index as a function of temperature was also carried out by means of the method of minimum deviation. A type K thermocouple was mounted inside a small hole drilled into the nontransmitting face of the prism in order to monitor the temperature of the prism. The thermocouple was held in place by Permatex Ultra Copper RTV silicone. The prism sample of $LiTbF_4$ was placed between two copper blocks, which were heated by two cartridge heaters within each block. The temperature was set using a Eurotherm 2416 temperature controller and allowed to stabilize for 45 min before refractive index data was taken. The temperature stability was ±1°C. Refractive indices were measured from 25 to 200 deg in increments of 25 deg.

3. RESULTS

The ordinary and extraordinary refractive indices of $LiTbF_4$ at their corresponding temperatures are shown in Figs. 1 and 2.

The data were fit to a modified version of a temperature-dependent Sellmeier equation discussed by Schlarb and Betzler [10] using the Levenburg–Marquardt algorithm,

$$n^2 = A + \frac{(B + CF)\lambda^2}{\lambda^2 - (\lambda_1 + DF)^2} + E\lambda^2, \tag{1}$$

where the parameter F is given by

$$F = (T - T_0) * (T + T_0 + 546.3). \tag{2}$$

The parameter T_0 in the expression for F represents the room temperature (taken as 23°C), and T is the temperature that the $LiTbF_4$ crystal was set at in order to take refractive index measurements. The additive factor of 546.3 represents the conversion of T and T_0 to the Kelvin scale. The values for the coefficients are shown in Table 1.

The values for dn/dT can be found by differentiating Eq. (1) in order to obtain

$$2n\frac{\partial n}{\partial T} = \left(\frac{(\lambda^2-(\lambda_1+DF)^2)C\lambda^2-((B+CF)\lambda^2)(2D(\lambda_1+DF))}{(\lambda^2-(\lambda_1+DF)^2)^2}\right)\frac{\partial F}{\partial T}. \tag{3}$$

The values of dn/dT at various wavelengths and temperatures are shown in Tables 2 and 3 and Fig. 3.

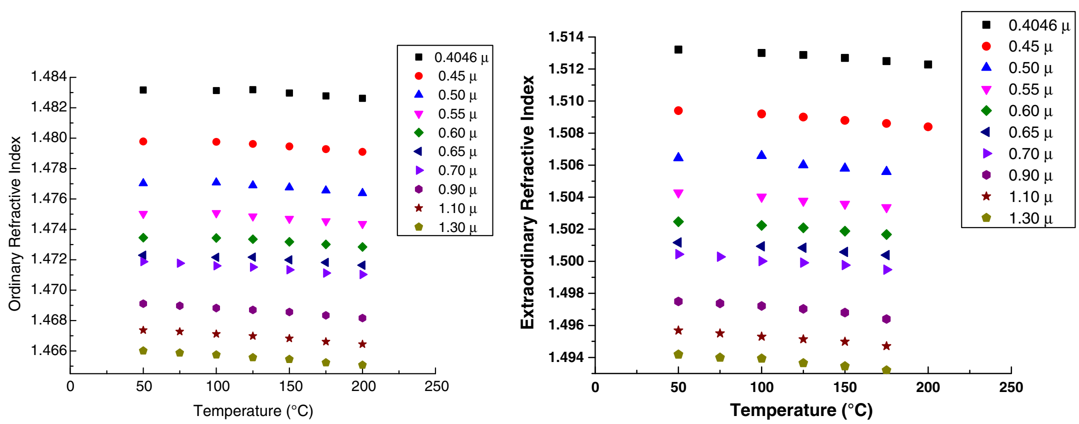

Fig. 2. Temperature dependence. (a) Ordinary refractive index. (b) Extraordinary refractive index.

Table 1. Temperature Dependent Sellmeier Coefficients for $LiTbF_4$

Sellmeier Parameter	n_o	n_e
A	1.80878	1.81294
B	0.34546	0.42458
C	-2.575×10^{-8}	-3.284×10^{-8}
D	1.924×10^{-8}	2.087×10^{-8}
E	−0.00497	−0.00518
λ_1	0.14003	0.13492

The data show that $LiTbF_4$ is positive birefringent with a negative dn/dT.

4. DISCUSSION

The effect of temperature on the optical elements used in high power laser systems has been studied extensively. [11] Output beams can be distorted due to nonuniformities in the temperature profile of the various elements in the system, and the performance of each of the elements can be compromised. In the case of magneto-optic devices such as Faraday isolators, the polarization of the output beam is affected by the temperature dependence of the Verdet coefficient and the addition of linear birefringence caused by mechanical stresses due to nonuniform temperature distribution through the elasto-optic effect.

Khazanov [12] has analyzed beam distortion and depolarization of beams in high power laser systems. In his analysis, the temperature dependent refractive index profile is expressed as [13]

$$n(r) = n(T_o) + [T(r) - T_o]P,$$

where

$$P = \frac{dn}{dT} - \alpha \frac{n_o^3}{4} \frac{1+\nu}{1-\nu}(p_{11} + p_{12}),$$

r is the radial distance from the center of the fiber, $\frac{dn}{dT}$ is the change of the refractive index with temperature, α is the thermal expansion coefficient, ν is the Poisson ratio, and p_{11} and p_{12} are the piezo-optic coefficients. In order to judge the utility of $LiTbF_4$ versus a more widely used material such as terbium gallium garnet (TGG), knowledge not only of dn/dT but also the thermal expansion coefficient, the Poisson ratio, and the piezo-optic constants is required. Regrettably, these measurements have not been made to sufficient accuracy in TGG [14], and to our knowledge, no such measurements have been performed on $LiTbF_4$. However, limited measurements of dn/dT have been measured for TGG, and the values are about twice those of $LiTbF_4$ and have the opposite sign. This may

Table 2. Values of dn_o/dT (×10⁶) Calculated from Eq. (3)

$\lambda(\mu)$	25°C	50°C	75°C	100°C	125°C	150°C	175°C	200°C	225°C
0.40	−8.78	−9.53	−10.3	−11.0	−11.8	−12.5	−13.3	−14.1	−14.8
0.50	−7.30	−7.92	−8.54	−9.16	−9.78	−10.4	−11.0	−11.7	−12.3
0.60	−6.59	−7.15	−7.71	−8.27	−8.82	−9.39	−9.95	−10.5	−11.1
0.70	−6.19	−6.72	−7.24	−7.76	−8.29	−8.81	−9.34	−9.86	−10.4
0.80	−5.95	−6.45	−6.95	−7.45	−7.95	−8.46	−8.96	−9.46	−9.97
0.90	−5.78	−6.27	−6.76	−7.24	−7.73	−8.22	−8.71	−9.20	−9.69
1.0	−5.67	−6.14	−6.62	−7.10	−7.58	−8.05	−8.53	−9.01	−9.49
1.1	−5.58	−6.05	−6.52	−6.99	−7.46	−7.93	−8.41	−8.88	−9.35
1.2	−5.52	−5.98	−6.45	−6.91	−7.38	−7.84	−8.31	−8.78	−9.24
1.3	−5.47	−5.93	−6.39	−6.85	−7.31	−7.78	−8.24	−8.70	−9.16
1.4	−5.44	−5.89	−6.35	−6.81	−7.26	−7.72	−8.18	−8.64	−9.10
1.5	−5.41	−5.86	−6.31	−6.77	−7.22	−7.68	−8.14	−8.59	−9.05
1.6	−5.38	−5.83	−6.29	−6.74	−7.19	−7.65	−8.10	−8.55	−9.01
1.7	−5.36	−5.81	−6.26	−6.72	−7.17	−7.62	−8.07	−8.52	−8.97

Table 3. Values of dn_e/dT (×10⁶) Calculated from Eq. (3)

$\lambda(\mu)$	25°C	50°C	75°C	100°C	125°C	150°C	175°C	200°C	225°C
0.40	−11.0	−12.0	−12.9	−13.9	−14.8	−15.8	−16.8	−17.7	−18.7
0.50	−9.21	−9.99	−10.8	−11.6	−12.3	−13.1	−13.9	−14.7	−15.5
0.60	−8.33	−9.03	−9.73	−10.4	−11.1	−11.9	−12.6	−13.3	−14.0
0.70	−7.82	−8.48	−9.15	−9.81	−10.5	−11.1	−11.8	−12.5	−13.1
0.80	−7.51	−8.14	−8.78	−9.41	−10.0	−10.7	−11.3	−12.0	−12.6
0.90	−7.30	−7.92	−8.53	−9.15	−9.77	−10.4	−11.0	−11.6	−12.2
1.0	−7.16	−7.76	−8.36	−8.97	−9.57	−10.2	−10.8	−11.4	−12.0
1.1	−7.05	−7.65	−8.24	−8.83	−9.43	−10.0	−10.6	−11.2	−11.8
1.2	−6.97	−7.56	−8.15	−8.73	−9.32	−9.91	−10.5	−11.1	−11.7
1.3	−6.91	−7.49	−8.08	−8.66	−9.24	−9.82	−10.4	−11.0	−11.6
1.4	−6.87	−7.44	−8.02	−8.60	−9.18	−9.75	−10.3	−10.9	−11.5
1.5	−6.83	−7.40	−7.98	−8.55	−9.13	−9.70	−10.3	−10.9	−11.4
1.6	−6.80	−7.37	−7.94	−8.51	−9.09	−9.66	−10.2	−10.8	−11.4
1.7	−6.77	−7.34	−7.91	−8.48	−9.05	−9.62	−10.2	−10.8	−11.3

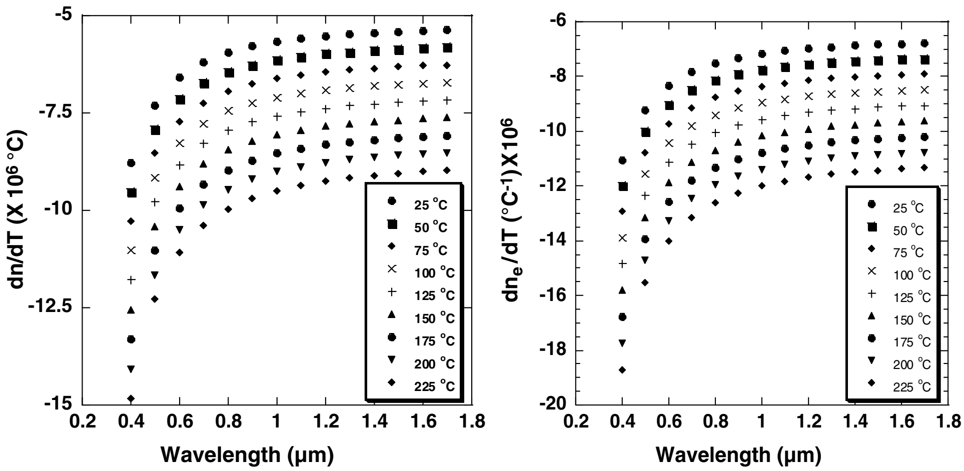

Fig. 3. Temperature and wavelength dependence of dn/dT: (a) dn_o/dT and (b) dn_e/dT.

make the problem of compensation for thermal lensing in isolators using $LiTbF_4$ more tractable.

5. CONCLUSION

We have measured the refractive index of $LiTbF_4$ as a function of wavelength and temperature. The parameters for a temperature dependent Sellmeier equation have been calculated and can be used for modeling the performance of $LiTbF_4$ in high power systems. The value obtained for dn/dT are about half those of TGG and are negative. This implies that the problem of compensating for thermal lensing for a Faraday isolator fabricated with $LiTbF_4$ should be simpler.

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